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Imaging XAFS study of simulated nuclear waste glass

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1 Introduction

High-level radioactive liquid waste (HLLW) generated in the nuclear fuel cycle is solidified with borosilicate glass in the melter. Behavior of fission product elements like Zr, Mo and Ru in the glass is very important information. The XAFS technique is very useful for analyzing chemical state of each element in the glass. In addition, we have used X-ray imaging technique to study behavior of some important elements like Ru during glass formation process [1, 2]. In the present work, distribution of each element was analyzed by using imaging XAFS technique.

2 Experiment

Imaging XAFS measurement was carried out at the NW10A beamline. The beam monitor AA40 and the high-sensitive CCD camera C11440-22C (Hamamatsu Photonics K.K.) was used to obtain 16bit grayscale images. Digital values in the image correspond to X-ray intensity for each pixels were used obtain XAFS spectrum[3]. It means that 1000 XAFS spectra can be obtained from digital grayscale values of 1000 pixels. The most remarkable feature in this method is that position sensitivity is added to the XAFS technique.

X-ray absorption images were recorded by X-ray energy scan passing through the absorption edge of Zr K (E_0=17.998keV), Mo K (E_0=20.000keV) and Ru K (E_0=22.117keV) for the same sample position. Distribution of each element was evaluated by analyzing absorption edge jump of each pixel in the image.

3 Results and Discussion

The X-ray absorption CCD image and distribution mapping results of Zr, Mo and Ru elements are shown in Fig.1. A black pixel in the mapping image means existence of each element. To the contrary, there is nothing for a white pixel. Many white circles found in the image are bubbles generated during denitration of the liquid waste. For Zr and Mo, they are uniformly distributed all over the glass sample except bubbles. On the other hand, a large aggregation was detected for Ru element. It is well known that operation in the Rokkasho plant was stopped by sediment of platinoids at the bottom of the melter in 2008. The imaging XAFS analysis used in the present work was effective in elucidation of the Ru behavior[2].

From imaging XAFS spectra, chemical state of each element was identified by XANES and EXAFS analyses. It can be estimated from XANES of Zr K-edge that the silicate like $ZrSiO_4$ is most likely for Zr element. The stable complex ion $(MoO_4)^{2-}$ was expected from curve fitting analysis of Mo-O pair. The chemical form of Ru element in the sediment was RuO_2. It is concluded that useful information on distribution and chemical form of each element in the glass can be obtained from the imaging XAFS technique.

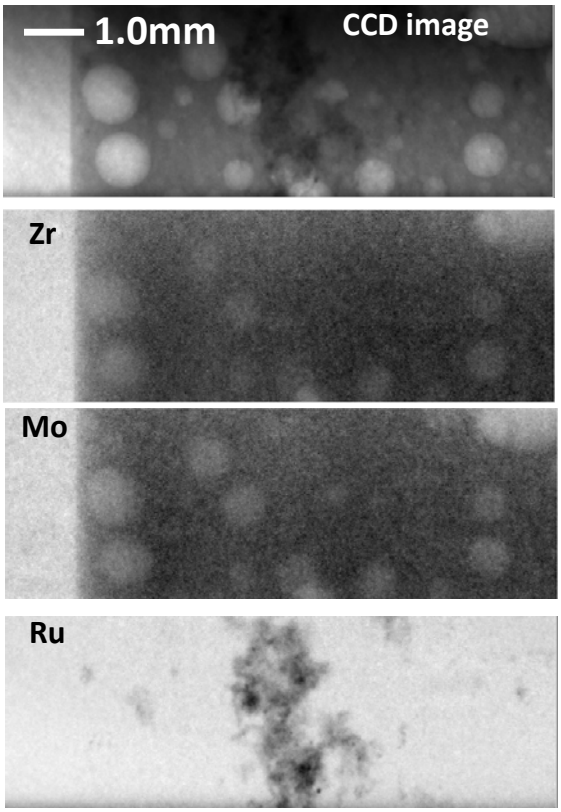

Fig. 1: X-ray absorption image (top) and distribution mapping images of Zr, Mo and Ru (lower three) in the simulated glass.

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References

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